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(71) Applicant (*for all designated States except US*): ATI PROPERTIES, INC. [US/US]; 2049 Century Park East, Los Angeles, CA 90067 (US).

(72) Inventors; and
(75) Inventors/Applicants (*for US only*): FROST, Chester, B. [US/US]; 1510 N.W. Woodland Drive, Corvallis, OR 97330 (US). KRUEGER, Brett, R. [US/US]; 36125 Providence School Road, Lebanon, OR 97355 (US).

(74) Agents: PUGH, Robert, J.; Allegheny Teledyne Incorporated, 1000 Six PPG Place, Pittsburgh, PA 15222 (US) et al.

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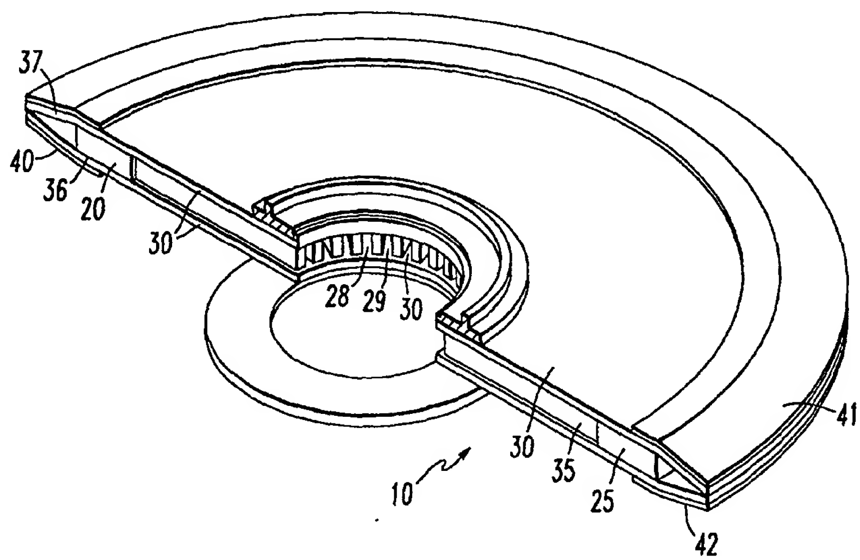
(54) Title: HYDROGEN SEPARATION MEMBRANE




(57) Abstract

A fluid separation assembly (110) having a fluid permeable membrane (138 and 162) and a wire mesh membrane (118 and 128) adjacent the fluid permeable membrane (138 and 162), wherein the wire mesh membrane (118 and 128) supports the fluid permeable membrane (138 and 162) and is coated with an intermetallic diffusion barrier. The barrier may be a thin film containing at least one of a nitride, oxide, boride, silicide, carbide and aluminide. Several fluid separation assemblies (110) can be used in a module (185) to separate hydrogen from a gas mixture containing hydrogen.

# HYDROGEN SEPARATION MEMBRANE

## BACKGROUND OF THE INVENTION

### 1. Field of the Invention

The present invention relates to apparatuses and methods for separation of a desired fluid from a fluid mixture. More particularly, the present invention is generally directed to a fluid separation assembly having a membrane permeable to a desired fluid and a wire mesh membrane support that supports the permeable membrane and has a barrier that prevents intermetallic diffusion bonding. This invention relates to metal membranes used for separating hydrogen gas from a mixture of gases.

### 2. Description of the Invention Background

Several metal foils have been shown to be suitable for the separation of hydrogen gas from a gas mixture by a mechanism of diffusion or transfer of hydrogen ions or protons through the normally non-porous metal foil with molecular hydrogen being recoverable on the opposite side of the foil from the side contacting the gas mixture. Many workers in this field have employed different metals, metal alloys and different structures to optimize the separation of the hydrogen component of a gas mixture or to purify hydrogen under the wide variety of conditions that can be present in a hydrogen containing gas stream at or near the conditions for hydrogen production. The U.S. Patents to Johann G.E. Cohn 3,238,700 and Leonard R. Rubin 3,172,742 are exemplary of the early work in this field which employed alloys of palladium to overcome some of the detrimental changes which can occur in the metal foil when subjected to substantial temperature changes in the presence of hydrogen, when produced typically by gas phase reactions such as the water gas shift reaction (steam over heated carbon or carbon containing materials), or the decomposition of hydrogen containing compounds. (Canadian Patent No. 579,535).

It is therefore an objective of the present invention to provide a supported hydrogen permeable metal or metal alloy foil-containing structure which can operate successfully in the environments present in hydrogen production as well as being

suitable for separation of hydrogen from gas streams containing hydrogen produced by any means.

Generally, when separating a gas from a mixture of gases by diffusion, the gas mixture is typically brought into contact with a nonporous membrane which is selectively permeable to the gas that is desired to be separated from the gas mixture. The desired gas diffuses through the permeable membrane and is separated from the other gas mixture. A pressure differential between opposite sides of the permeable membrane is usually created such that the diffusion process proceeds more effectively, wherein a higher partial pressure of the gas to be separated is maintained on the gas mixture side of the permeable membrane. It is also desireable for the gas mixture and the selectively permeable membrane to be maintained at elevated temperatures to facilitate the separation of the desired gas from the gas mixture. This type of process can be used to separate hydrogen from a gas mixture containing hydrogen. Thus, in this application, the permeable membrane is permeable to hydrogen and is commonly constructed from palladium or a palladium alloy. The exposure to high temperatures and mechanical stresses created by the pressure differential dictates that the permeable membrane be supported in such a way that does not obstruct passage of the desired gas through the membrane.

One type of conventional apparatus used for the separation of hydrogen from a gas mixture employs a woven refractory-type cloth for supporting the permeable membrane during the separation process. The disadvantage of this type of conventional membrane support is that the cloth support is susceptible to failure when it is exposed to high mechanical stresses associated with the differential pressure required to effect diffusion through the membrane material.

Another conventional permeable membrane support is a metal gauze structure placed adjacent to the permeable membrane. The disadvantage of this type of support is that intermetallic diffusion bonding occurs between the membrane support and the permeable membrane when they are exposed to high pressures and high temperatures. The high pressure tends to compress the permeable membrane and the metal gauze together and the high temperatures tend to deteriorate the chemical bonds of those materials. Such undesirable condition results in migration of the molecules of the permeable membrane to the metal gauze membrane and the migration of molecules of

the metal gauze membrane to the permeable membrane until a bond is formed between those two structures. This intermetallic diffusion bonding results in a composite material that is no longer permeable by the hydrogen gas.

Thus, the need exists for a method and apparatus for separating a desired fluid from a fluid mixture that can reliably withstand high operating pressures and temperatures.

Another need exists for a permeable membrane and support arrangement for separating a desired fluid from a fluid mixture, wherein the permeable membrane is not susceptible to breakage or intermetallic diffusion bonding.

Yet another need exists for a method of supporting a membrane that is permeable to a fluid, wherein the fluid permeable membrane is exposed to high temperatures and high pressures.

## BRIEF DESCRIPTION OF THE DRAWINGS

For the present invention to be readily understood and practiced, preferred embodiments will be described in conjunction with the following figures wherein:

FIG. 1 is an isometric view of a membrane assembly according to the present invention before welding;

FIG. 2 is a top view of the membrane assembly of FIG. 1;

FIG. 2A is a sectional view of the membrane assembly of FIG. 2 taken along the lines and arrows 2A;

FIG. 3 is a partial sectional view in perspective of the edge detail of the membrane of the present invention;

FIG. 4 is a partial cut away view of the membrane assembly of the present invention;

FIG. 5 is a top isometric view of a fluid separation assembly of the present invention;

FIG. 6 is an exploded isometric view of the fluid separation assembly of the present invention shown in FIG. 5;

FIG. 7 is an exploded isometric view of the female permeable membrane subassembly of the present invention shown in FIG. 5;

FIG. 8 is an exploded isometric view of the male permeable membrane subassembly of the present invention shown in FIG. 5;

FIG. 9 is a sectional isometric view of the fluid separation assembly of the present invention;

FIG. 10 is an enlarged view of section **A** of the fluid separation assembly shown in FIG. 9;

FIG. 11 is a cross-sectional view of the fluid separation assembly of the present invention shown in FIG. 5 taken along line 11-11;

FIG. 12 is an isometric sectional diagrammatical view of a module employing several fluid separation assemblies of the present invention; and

FIG. 13 is an enlarged section **B** of the module shown in FIG. 12.

## BRIEF SUMMARY OF THE INVENTION

The hydrogen permeable membrane structure of the present invention comprises a disc shaped supported metal foil sized to overlap a support plate or disc capable of passing product gases diffused or transferred through the metal foil, which disc terminates at its periphery with a pair of beveled sheet metal support rings overlying the support plate and a pair of beveled sheet metal capture rings matched in shape to the beveled support rings to capture the hydrogen permeable membrane foil there between, the assembly terminating in an edge formed by the edges of the capture rings and support rings with the captured foil, which edge is then hermetically sealed by a weld bead joining all of the rings together. The assembly includes means for centrally removing the separated hydrogen from the support plate.

The present invention provides a fluid separation assembly having a fluid permeable membrane and a wire mesh membrane support adjacent the fluid permeable membrane, wherein the wire mesh membrane support has an intermetallic diffusion bonding barrier.

The present invention further provides a method for separating a desired fluid from a fluid mixture comprising a membrane that is permeable by the desired fluid, providing a wire mesh membrane support with a intermetallic diffusion bonding barrier, wherein the wire mesh membrane support is adjacent to the fluid permeable

membrane, contacting the fluid permeable membrane support with the fluid mixture and contacting the wire mesh membrane support with the desired fluid permeating the fluid permeable membrane.

The present invention further provides for a method of making a fluid separation assembly comprising providing a membrane permeable to a desired fluid, providing a first retainer, providing a wire mesh membrane support having an intermetallic diffusion bonding barrier and placing it adjacent the fluid permeable membrane, providing a permeate member adjacent the wire mesh membrane support, providing a gasket adjacent the fluid permeable membrane, providing a second retainer adjacent the wire mesh membrane support and joining the first retainer, the gasket and the second retainer at their peripheries.

The present invention provides for a method for supporting a fluid permeable membrane comprising providing a membrane that is permeable by a desired fluid, and providing a wire mesh membrane support with an intermetallic diffusion bonding barrier, wherein the wire mesh membrane support is adjacent and supports the fluid permeable membrane.

Other details, objects and advantages of the present invention will become more apparent with the following description of the present invention.

## DETAILED DESCRIPTION

The present invention will be described below in terms of apparatuses and methods for separation of hydrogen from a mixture of gases. It should be noted that describing the present invention in terms of a hydrogen separation assembly is for illustrative purposes and the advantages of the present invention may be realized using other structures and technologies that have a need for such apparatuses and methods for separation of a desired fluid from a fluid mixture containing the desired fluid.

It is to be further understood that the Figures and descriptions of the present invention have been simplified to illustrate elements that are relevant for a clear understanding of the present invention, while eliminating, for purposes of clarity, other elements and/or descriptions thereof found in a hydrogen separation assembly. Those of ordinary skill in the art will recognize that other elements may be desirable

in order to implement the present invention. However, because such elements are well known in the art, and because they do not facilitate a better understanding of the present invention, a discussion of such elements is not provided herein.

In the manufacture of a metal membrane based gas separation device, it is necessary to eliminate all possible leak paths between the feed side of the membrane and the product or permeate side of the membrane. This is done to maintain the highest possible purity of the product stream. Typically, a membrane used in this type of a device is very thin and consequently of low mechanical strength. A mechanical support, placed under the membrane on the product side, allows the membrane to function under high differential pressures at elevated temperatures. If membrane material is placed on both sides of the mechanical support, then the support itself is loaded under compressive stress during operation. Using this type of a design, the mechanical support can be less robust than would be necessary if it were loaded form one side only.

The present invention provides a way of sealing each membrane "half" independent of the mechanical support so that all possible leak paths are eliminated. If both membrane sheets are joined at their periphery using a single seam type weld to join parts, the probability of a leak through a welded seam is substantially reduced.

Many methods of the joining of thin metal foils used for hydrogen permeable membranes are known. Efforts to achieve a hermetic seal of thin foil type membranes with welding, brazing or diffusion bonding are described in U.S. Patent No. 5,645,626. Additional efforts at a brazed joint are described in Canadian Patent Application Number 436,620. The problems with a brazed type joint occur because of the possibility of the brazing alloy contaminating the membrane surface during the brazing operation with metals, flux compounds, or other substances that would interfere with the membranes ability to transport hydrogen during operation. Diffusion bonding of membrane foils as a means of attachment to fabricated support members is limited by the requirement that a hermetic seal in a bonded join can only occur when the material in the bonding zone enters into the liquid stage at some time during the bonding process.

The present invention involves the joining of two thin foil membranes (approx. 25 micron/0.001 inch) using commercially available welding technology. A

single weld joint hermetically seals both membranes in one pass, reducing the probability of a failed weld joint due to weld induced stress, contamination present in the base metals, and of porosity in the finished weld by approximately 50%. The completed joint has advantages over present technology in the areas of reliability and ease of manufacturing.

The hydrogen separation membrane of the present invention is therefore an improvement over the membrane structure described in U.S. Patent No. 5,139,541 whose description is incorporated herein by reference in its entirety.

The membrane support structure 10 of the present invention is, as shown in Fig. 1, a disc shaped member designed to contact the hydrogen containing gas with both of its exterior surfaces. The exposed exterior surface 12 of the foil is sandwiched under the outer ring 13 in a manner more fully described hereinafter. The central hole communicates with the interior space of the disc for removal of the hydrogen. The reverse side of the disc (not shown) is identical as can be seen on Figs. 2 and 2A. The internal support disc has a hole corresponding to the interior of the central support structure 21. The disc 20 has a solid perimeter which is shown in section at 25. Slots are provided in the disc which radiate radially outward from its central hole to terminate at the edge of the outer perimeter. The edges of the slots are shown schematically at 27, 28 and 29 for example. The foil 30 is received on both faces of the disc 10. Hydrogen will be transported from the exterior of the disc 10 through the foil 30 and into a slot 35 in the support disc from where it can be removed through a central conduit (not shown) since the edges of the slots (for example 28, 29 and 30) communicate with the opening (hole 15) and the central conduit. Other means for permitting the exhaust of the hydrogen can be provided i.e. such as grooves instead of slots or surface texturing or the like.

As previously described, the edge structure for the disc is critical for achieving a gas tight seal. Likewise, the foil preferably will be supported on an inert fabric such as woven fiberglass or a woven or non-woven ceramic material. Referring now to Fig. 3, the support disc 20 is covered on both sides with a layer of inert fabric 31. The hydrogen permeable metal membrane foil 30 is received over the fabric 30. A pair of sheet metal support rings 36 and 37 are affixed to the support disc 20 around its periphery so as not to occlude the slots. The beveled support rings 36 and 37 are

dimensional to meet and form an edge. The foil 30 and 32 are positioned to overlay the fabric 31 received on the fabric 31 supported by the support disc 20. A pair of sheet metal capture rings 40 and 41 are sized to mate with the support rings 36 and 37 respectively and capture the foil 30 and 32 between the support rings 36 and 37 and the metal capture rings 40 and 41. A weld bead 50 joins all of the rings together. The disc 20, capture rings 40 and 41, and support rings 36 and 37 are all made from materials which are compatible with the end use environment for the membrane discs.

This invention differs from the present technology by reducing the total length of welded or sealed joints for a given pair of membranes with comparable outside diameters. The invention also allows dimensional changes due to the different coefficients of thermal expansion used in the materials of the welded joint and dimensional changes encountered when the membrane material is saturated with hydrogen which occurs independently of other components in the assembly. This has the effect of further reducing stress on the membrane under operating conditions. Additionally, this type of joint allows for the selection of lower cost materials of construction that are not physically a part of the welded joint further reducing the cost of manufacture.

The fabrication of these parts into a completed assembly is accomplished by assembly into a welding fixture that clamps the parts together. The membrane material is mechanically forced between the bottom membrane support and the membrane cover rings during clamping into the weld fixture so that the outer edges are held tightly together in tight contact with the membrane. This clamped assembly is then seam welded at the outer perimeter. The single weld bead fuses the outer edges of the upper and lower bottom membrane support rings, the upper and lower membranes, and the upper and lower membrane cover rings into a leak proof unit.

The actual seam weld can be effected by a number of commercially available technologies, including but not limited to laser, electron beam, and tungsten inert gas (TIG) welding. The welded seam can be achieved with or without the addition of filler material to the weld zone. This type of weld may have other applications requiring welding of thin metal foils for other uses outside the membrane application described.

An assembly of forty-three individual discs prepared as described herein, containing approximately twenty square feet of total membrane area were operated at 300° C and 600 psig for a total of 200 hours with no noticeable decline in hydrogen separation function. The permeate side of the assembly was operated at a pressure of from about 15 to 30 psig subjecting the assembly to a differential pressure across each membrane of 570-585 psig. The assembly was also subjected to about 30 cycles of operation from ambient conditions of temperature and pressure to the operating temperatures and pressures described without failure of the welded joint.

The limitations to maintaining weld integrity and thus a hermetically sealed weld joint are primarily in the selection of materials for the bottom support and top cover rings that are compatible with the thin metal foil to be welded and selecting the proper weld parameters of the equipment used to make the weld.

FIGS. 5 and 6 illustrate one embodiment of the fluid separation assembly 110 of the present invention, wherein FIG. 6 is an exploded view of the fluid separation assembly 110 shown in FIG. 5. The fluid separation assembly 110 comprises first membrane retainers 112, a female membrane subassembly 114, a first membrane gasket 116, a first wire mesh membrane support 118, second membrane retainers 120, a slotted permeate plate 122, a permeate rim 124, a second wire mesh membrane support 128, a second membrane gasket 130 and a male membrane subassembly 132. In one embodiment, the first retainers 112 may be substantially flat ring members having an outside diameter equal to the diameter of the female and male membrane subassemblies 114 and 132 and a thickness of between approximately 0.001 inches and 0.060 inches. The first membrane retainers 112 each have a centrally disposed opening 113 and 135. The first membrane retainers 112 may be made from Monel 400 (UNS N 04400); however, other materials that are compatible with the welding process, discussed below, may also be used. It will also be appreciated that while first retainers 112 are shown as comprising substantially annular members they may have other desired shapes and other thicknesses without departing from the spirit and scope of the present invention.

FIG. 7 is an exploded view of a female permeable membrane subassembly 114. In this embodiment, female membrane subassembly 114, comprises a female gasket seat 136, a hydrogen permeable membrane 138, an inner diameter membrane

gasket 140 and a center support washer 142. In this embodiment, the female gasket seat 136 is a substantially flat ring member 144 having a raised face 146 extending around the ring member 144 and a centrally disposed opening 145. It will be appreciated that while this embodiment is shown with gasket seats with this configuration, there may be other geometries of gasket seats specific to other gasket configurations or materials that may be used without departing from the spirit and the scope of the present invention. The female gasket seat 136 may be made from Monel 400; however, other materials such as nickel, copper, nickel alloys, copper alloys, or other alloys that provide for compatible fusion with the chosen permeable membrane material during welding may be used. In this embodiment, the hydrogen permeable membrane 138 is a substantially planar member having a circular configuration, opposing sides 148 and a centrally disposed circular opening 150. The inner diameter membrane gasket 140 is also a flat ring member having a centrally disposed opening 151. Also in this embodiment, the inner diameter membrane gasket 140 may be made from Monel 400 (UNS N 04400); however, other materials such as nickel, copper, nickel alloys, copper alloys, or other alloys that provide for compatible fusion with the chosen permeable membrane material during welding may be used. The center support washer 142 is a flat ring member having a centrally disposed opening 153. The center support washer 142 may be made of Monel 400 (UNS N 04400); however, other materials such as nickel, copper, nickel alloys, copper alloys, or other alloys that provide for compatible fusion with the chosen permeable membrane material or alloy during welding may be used.

Referring back to FIG. 6, in this embodiment, the first and second membrane gaskets 116 and 130 are each a substantially flat ring member having a centrally-disposed opening 155 and 157, respectively. In this embodiment, the first and second membrane gaskets 116 and 130 may be made from Monel 400 alloy (UNS N 004400), nickel, copper, nickel alloys, copper alloys or other precious alloys or other alloys compatible with the weld that is used to join the components of the fluid separation assembly 110 and which is discussed below. The first and second membrane gaskets 116 and 130 may have a thickness of between approximately 0.0005 inches to 0.005 inches. However, other gasket thicknesses could be employed.

Also in this embodiment, the first and second wire mesh membrane supports 118 and 128 are planar, ring-shaped members having centrally disposed openings 152 and 154, respectively. The wire mesh membrane supports 118 and 128 may be made from 316L stainless steel alloy with a mesh count of between approximately 19 to 1,000 mesh per inch, wherein the mesh count is chosen to be adequate to support the hydrogen permeable membranes 138 and 162. The style of woven mesh may include a standard plain square weave, twill square weave, rectangular plain or twill weave or triangular plain or twill weave. One example of a mesh count that may be used is 49 mesh per inch. The wire mesh membrane supports 118 and 128 may be made of steel alloys, stainless steel alloys, nickel alloys or copper alloys. The wire mesh may be coated with a thin film that prevents intermetallic diffusion bonding (i.e., an intermetallic diffusion bonding barrier). The intermetallic diffusion bonding barrier may be a thin film containing at least one of an oxide, a nitride, a boride, a silicide, a carbide, or an aluminide and may be applied using a number of conventional methods, including but not limited to, physical vapor deposition (PVD), chemical vapor disposition, and plasma enhanced vapor deposition. For example, the method of reactive sputtering, a form of PVD, can be used to apply a thin oxide film of between approximately 600-700 angstroms to the wire mesh membrane supports 118 and 128. A variety of oxides, nitrides, borides, silicides, carbides and aluminides may also be used for the thin film as well as any thin films that will be apparent to those of ordinary skill in the art. Using this form of PVD results in a dense amorphous thin film having approximately the same mechanical strength as the bulk thin film material.

Also in this embodiment, the second membrane retainers 120 each are a substantially flat ring member. One retainer 120 has a centrally disposed opening 159 and retainer 120 has a centrally disposed opening 161. See FIG. 6. These retainers 120 may be the same thickness as the first and second wire mesh membrane supports 118 and 128. The second membrane retainers 120 may be made from a material that is compatible with the weld, discussed below, such as Monel 400 (UNS N 004400) and nickel, copper, nickel alloys, eopper alloys, precious metals or alloys, or other alloys that provide for compatible fusion with the chosen membrane material or alloy during welding may be used.

In this embodiment, the slotted permeate plate 122 is a steel plate having a plurality of slots 156 extending radially and outwardly from a central opening 158 in the direction of the periphery of the slotted permeate plate 122. The number of slots 156 in a slotted permeate plate 122 may range from approximately 10 to 72. However, other suitable slot densities could conceivably be employed. The permeate plate rim 124 is a substantially flat ring member having a centrally disposed opening 163 and an inner diameter larger than the outer diameter of the slotted permeate plate 122. The permeate plate rim 124 is made from Monel 400 (UNS N 04400); however, other materials can also be used such as nickel, copper, nickel alloys, copper alloys, precious metals or alloys or other alloys that provide for compatible fusion with the chosen membrane material or alloy during welding.

FIG. 8 is an exploded view of the male permeable membrane subassembly 132. The male membrane subassembly 132 comprises a male gasket seat 160, a hydrogen permeable membrane 162, an inner diameter membrane gasket 164, and a center support washer 166. The hydrogen permeable membranes 138 and 162 may be made from at least one hydrogen permeable metal or an alloy containing at least one hydrogen permeable metal, preferably selected from the transition metals of groups VIIB or VIIB of the Periodic Table. The hydrogen permeable membrane 162, the inner diameter membrane gasket 164, and the center support washer 166 are similar in structure to the hydrogen permeable membrane 138, the inner diameter membrane gasket 140 and the center support washer 142, respectively, discussed above. The male gasket seat 160 is a substantially planar ring member 168 having a circular protuberance 170 extending around a centrally disposed opening 172. In this embodiment, the female gasket seat 136 and the male gasket seat 160 are made of a high strength alloy material that is compatible with the weld such as Monel 400. The inner diameter member gaskets 140 and 164 are made from the same materials as the first and second outer diameter membrane gaskets 116 and 130, discussed above.

FIGS. 9 through 11 are various cross-sectional views of the assembled fluid separation assembly 110 of the present invention, wherein FIG. 10 is an enlarged view of section A of the fluid separation assembly 110 shown in FIG. 9, and FIG. 11 is a cross-sectional plan view of the assembled fluid separation assembly 110. When assembling the components of the fluid separation assembly 110 shown in FIGS. 6-8,

the female membrane subassembly 114 and the male membrane subassembly 132 are initially assembled. The female gasket seat 136, the permeable membrane 138, the inner diameter membrane gasket 140 and the center support washer 142 are placed adjacent one another, as shown in FIG. 11, such that their central disposed openings 145, 150, 151 and 153, respectively, are coaxially aligned. A first weld 171, shown in FIG. 11, is placed at the openings thereof. The first weld 171 takes the form of a weld bead creating a hermetic seal between the female gasket seat 136, the permeable membrane 138, the inner diameter membrane gasket 140 and the center support washer 142. The weld 171 can be effected by a number of commercially available technologies, including but not limited to, lasers, electron beam and tungsten inert gas (TIG) welding. Alternative joining technologies such as brazing or soldering may also be employed with the desired result being a gas tight bond between the gasket seat 136 and the permeable membrane 138. Likewise, the components of the male membrane subassembly 132, which include the male gasket seat 160, the permeable membrane 162, the inner diameter membrane gasket 166 and the center support washer 166 are also placed adjacent one another, as shown in FIG. 11, such that their centrally disposed openings 172, 181, 183 and 185 are coaxidly aligned with each other and a second weld bead 173, shown in FIG. 11, is placed around the circumference of the openings 172, 181, 183 and 185 thereof. As stated above, the weld 173 can be effected by a number of commercially available technologies, including but not limited to, laser, electron beam, and tungsten inert gas (TIG) welding.

After the components of the female membrane subassembly 114 and the components of the male membrane subassembly 132 have each been connected by the welds 171 and 173, respectively, they are assembled with the other components described above to form the fluid separation assembly 110. As shown in FIG. 6, the first and second retainer members 112 and 120, the female and male membrane subassemblies 114 and 132, the first and second outer diameter gaskets 116 and 130, the first and second wire mesh membrane supports 118 and 128, the slotted permeate plate 122 and the permeate rim 124 are aligned such that their centrally disposed openings are coaxially aligned. As shown in FIG. 11, these components are retained in that configuration by placing a weld 174 at the outer periphery of the first and

second retainer members 112 and 120, the female and male membrane subassemblies 114 and 132, the first and second outer diameter membrane gaskets 116 and 130, and the slotted permeate rim 124. Alternatively, these parts could be assembled such that their centrally disposed openings are coaxially aligned, as shown in FIG. 11, and connected to one another by performing a brazing or soldering operation at the outer periphery of the first and second retainer members 112 and 120, the female and male membrane subassemblies 114 and 132, the first and second outer diameter membrane gaskets 116 and 130 and the slotted permeate rim 124. As seen in FIG. 10, a space 175 is provided between the slotted permeate plate 122 and the permeate rim 124 which permits expansion and contraction of the components of the fluid separation assembly 110 resulting from the change in temperature. Assembled, the fluid separation assembly 110 may have a thickness ranging from 0.010 inches to 0.125 inches, depending upon the thicknesses of the components employed.

When separating the hydrogen from a mixture of gas that includes hydrogen, the gas mixture is directed towards the permeable membranes 138 and 162 of the female membrane subassembly 114 and the male membrane subassembly 132, respectively, in the directions **D** and **E**, as shown in FIG. 11. For clarity, the permeable membranes 138 and 164 of the female and male membrane subassemblies 114 and 132, respectively are shown in FIG. 11 as being spaced from the first and second wire mesh membrane supports 118 and 128; however, in use, the permeable membranes 138 and 162 are in contact with the first and second wire mesh membrane supports 118 and 128 and are supported thereby. When the gas mixture containing hydrogen contacts the hydrogen permeable membranes 138 and 162, the hydrogen permeates through the permeable membranes 138 and 162, passes through the first and second wire mesh membrane supports 118 and 128 and enters the slotted permeate plate 122 where the hydrogen enters a specific slot 156 and to be directed toward the central axis **C** by the passageways formed by the slots 156. The central openings of the components of the fluid separation assembly 110, shown in FIG. 6, form a conduit 180 wherein the purified hydrogen is collected and transported to a desired location. The conduit 180 may have a diameter of between approximately 0.25 inches and 1 inch. The diameter is determined by the components of the fluid separation assembly 110 and by the desire that the hydrogen flow be substantially

unimpeded. The non-hydrogen gases in the gas mixture are prevented from entering the fluid separation assembly 110 by the fluid permeable membranes 138 and 162. The remainder of the hydrogen depleted gas mixture is directed around the exterior of the fluid separation assembly 110 in this embodiment.

FIGS. 12 and 13 illustrate a module 185 employing several fluid separation assemblies 110 of the present invention, wherein FIG. 13 is an enlarged section **B** of the module 185. Each of the fluid separation assemblies 110 are shown as a solid body for clarity. However, each of the fluid separation assemblies 110 are the same as the fluid separation assemblies 110 shown in FIGS. 5-11. The module 185 has a feed gas inlet 191, a permeate outlet 190 and a discharge gas outlet 193. The fluid separation assemblies 110 are coaxially aligned. Distribution plates 187 are sandwiched between and separate the fluid separation assemblies 110. The distribution plates 187 are positioned on a shoulder of the gasket seats 136 in such a manner that they are positioned equidistant from the planar surface of the permeable membrane assemblies 114 and 132 in successive fluid separation assemblies 110. The distribution plates 187 are not fixedly connected to the gasket seats 136 and 160, but rather rest on a shoulder of the gasket seat 136. There is sufficient clearance between the central opening of the redistribution plate 187 and the shoulder on the female gasket seat 136 that the redistribution plates 187 and the fluid separation assemblies 110 are allowed to position themselves inside the wall of the membrane housing independently of the position of the fluid separation assemblies 110. Each distribution plate 187 has openings 189 therein. The fluid separation assemblies 110 are aligned one with the other such that each of the conduits 180 of the fluid separation assemblies 110 form a larger conduit 190. The path of the gas mixture containing hydrogen, represented by arrow **G**, enters the opening 189 and travels along the outer surface of the fluid separation assembly 110, wherein some of the hydrogen of the gas mixture is free to enter the fluid separation assembly 110 by the permeable membranes 138 and 162 and is directed along path **F** into the larger conduit 190 and the remaining gas mixture follows arrow **G** and serpentines through the passageway, formed by the distribution plates 187, the fluid separation assemblies 110 and the interior wall 192 of the module 185. As the gas mixture travels through the passageway, it contacts the outer surfaces of several other fluid separation

assemblies 110, wherein more of the hydrogen remaining in the gas mixture permeates the permeable membrane 138 and 162 and follows the path F resulting in this purified hydrogen entering the larger conduit 190. The remainder of the hydrogen depleted gas mixture exits through a port 193 located at the opposite end of the module 185 after flowing over the entire stack of fluid separation membrane assemblies 110.

Although the present invention has been described in conjunction with the above described embodiments thereof, it is expected that many modifications and variations will be developed. This disclosure and the following claims are intended to cover all such modifications and variations

WHAT IS CLAIMED:

1. A hydrogen permeable membrane separation module comprising a perforate disc having an imperforate band circumferential of the disc and at the outer edge thereof and having a top surface and a bottom surface and voids which communicate with a collection chamber, located at the center of the disc, two performate inorganic barriers overlying said perforate disc on both the top and bottom surfaces, two hydrogen permeable metal membranes overlaying said perforate inorganic barrier and terminating beyond the outer circumference of the perforate disc, two circular support rings overlaying the imperforate band of said disc, said barrier terminating on the disc short of the outer circumference thereof and abutting the circular support rings, the outer circumference of said membrane received on the support rings, and capture rings received on the membranes and sized to terminate circumferentially with the membranes and support rings and a weld bead connecting the outer capture rings, the membranes and the support rings in a fluid tight manner so that hydrogen can pass through the membranes, the barrier and the voids in the perforate disc and be collected centrally of the assembly.

2. A fluid separation assembly, comprising:
a fluid permeable membrane; and
a wire mesh membrane adjacent said fluid permeable membrane, said wire mesh membrane having an intermetallic diffusion barrier.

3. The fluid separation assembly according to claim 2, wherein said barrier is a thin film containing at least one of one of the group consisting of nitrides, oxides, borides, silicides, carbides and aluminides.

4. The fluid separation assembly according to claim 3, wherein said barrier is a thin film containing one of an oxide and a nitride.

5. The fluid separation assembly according to claim 2, wherein said wire mesh membrane is in contact with said fluid permeable membrane.

6. The fluid separation assembly according to claim 2, wherein said fluid permeable membrane is a substantially planar member having a centrally disposed opening.

7. The fluid separation assembly according to claim 6, wherein said wire mesh membrane is a substantially planar membrane having a centrally disposed opening which is in alignment with said fluid permeable membrane opening.

8. The fluid separation assembly according to claim 7, wherein said wire mesh membrane has a mesh count ranging between approximately 19 to 1000 mesh per inch.

9. The fluid separation assembly according to claim 7, further comprising a slotted permeate plate adjacent to said wire mesh membrane.

10. The fluid separation assembly according to claim 9, further comprising a second fluid permeable membrane and a second wire mesh membrane, wherein said slotted permeate plate has a first side and a second side and said first permeable membrane is adjacent said first side of said slotted permeate plate and said first wire mesh membrane is adjacent said first permeable membrane, and wherein said second wire mesh membrane is adjacent said slotted permeate plate second side and said second fluid permeable membrane is adjacent said second wire mesh membrane.

11. The fluid separation assembly according to claim 10, wherein said slotted permeate plate, said second wire mesh membrane and said second fluid permeable membrane each also have a centrally disposed opening and each of said centrally disposed openings are coaxially aligned and form a central conduit.

12. The fluid separation assembly according to claim 2, wherein said wire mesh membrane is made from stainless steel.

13. The fluid separation assembly according to claim 10, wherein each of said fluid permeable membranes further comprises a gasket seat, a membrane gasket, and a washer to form a first and second membrane subassembly, wherein said gasket seats, said membrane gaskets and said washers are connected to said fluid permeable membranes.

14. The fluid separation assembly according to claim 13, further comprising a weld bead connected to each of said first and second membrane subassemblies.

15. The fluid separation assembly according to claim 14, further comprising first retainers, one of said first retainers connected to each of said fluid permeable membranes.

16. The fluid separation assembly according to claim 14, further comprising second retainers adjacent said slotted permeate plate.

17. The fluid separation assembly according to claim 14, further comprising first retainers, one of said second retainers adjacent each of said fluid permeable membranes.

18. The fluid separation assembly according to claim 14, further comprising gaskets, one of said gaskets adjacent each of said wire mesh membranes.

19. A fluid separation assembly, comprising:

a slotted permeate having opposing faces;

first and second wire mesh membranes, each of said wire mesh membranes having a first surface and a second surface, wherein each of said wire mesh membranes first surfaces are adjacent said slotted permeate;

first and second membranes permeable to a desired fluid, each of said permeable membranes adjacent one of said wire mesh membranes second surfaces;

a permeate rim surrounding said slotted permeate;

first retainers adjacent each of said permeable membranes;

second retainers adjacent said slotted permeate and said wire mesh membranes; and

gaskets between each of said wire mesh membranes and said permeable membranes, wherein said permeate rim, said first retainers, said second retainers said permeable membranes and said gaskets are joined together at their peripheries.

20. The fluid separation assembly according to claim 19, wherein said permeate rim, said first retainers, said second retainers, said permeable membranes and said gaskets are jointed together by a weld bead at their peripheries.

21. The fluid separation assembly according to claim 19, further comprising a female gasket seat, a membrane gasket and a washer, wherein said female gasket seat, said membrane gasket and said washer are connected to one of said permeable membranes and comprise a female membrane subassembly.

22. The fluid separation assembly according to claim 21, further comprising a male gasket seat, a second membrane gasket, and a second washer, wherein said male gasket seat, said second membrane gasket and said second washer are connected to the other of said fluid permeable membranes and comprises a male membrane subassembly.

23. The fluid separation assembly according to claim 22, wherein each of said gasket seats, said membrane gaskets, said washers and said permeable membranes have a centrally disposed opening and said openings are coaxially aligned and first and second weld beads connect the components of each subassembly.

24. The fluid separation assembly according to claim 19, wherein each of said two wire mesh membranes have an intermetallic diffusion bonding barrier.

25. The fluid separation assembly according to claim 24, wherein said intermetallic diffusion bonding barrier is a thin film containing at least one of the group consisting of oxides, nitrides, borides, silicides, carbides and aliminides.

26. The fluid separation assembly according to claim 19, wherein said first retainers, said second retainers, said gaskets, said permeate rim and said two membranes are connected at their peripheries.

27. The fluid separation assembly according to claim 26, wherein a weld bead is located at said peripheries of each of said first retainers, said second retainers, said gaskets, said permeate rim and said two membranes.

28. The fluid separation assembly according to claim 19, wherein each of said two wire mesh membranes are stainless steel.

29. The fluid separation assembly according to claim 19, wherein each of said two wire mesh membranes have mesh counts ranging from approximately 19 to 1000 mesh per inch.

30. The fluid separation assembly according to claim 29, wherein each of said two wire mesh membranes have mesh counts ranging from 49 to 1000 mesh per inch.

31. The fluid separation assembly according to claim 27, wherein each of said permeable membranes and said slotted permeate have a centrally disposed opening that form a conduit.

32. A fluid separation module, comprising:

a plurality of fluid separation assemblies, wherein each of said fluid separation assemblies comprises:

a slotted permeate having opposing faces;

first and second wire mesh membranes, each of said wire mesh membranes having a first surface and a second surface, wherein each of said wire mesh membranes first surfaces is adjacent said slotted permeate;

first and second membranes permeable to a desired fluid, each of said permeable membranes adjacent one of said wire mesh membranes second surfaces;

a permeate rim surrounding said slotted permeate;

first retainers adjacent each of said permeable membranes;

second retainers between said slotted permeate and each of said wire mesh membranes; and

gaskets between each of said wire mesh membranes and permeable membranes, wherein said permeate rim, said first retainers, said second retainers and said gaskets are joined together at their peripheries.

33. A method for separating a desired fluid from a fluid mixture, comprising:

providing a membrane that is permeable by the desired fluid and having opposing surfaces;

providing a wire mesh membrane with an intermetallic diffusion bonding barrier, wherein the wire mesh membrane is adjacent to one of the opposing surfaces of the fluid permeable membrane;

contacting the fluid permeable membrane with the fluid mixture; and

contacting the wire mesh membrane with the desired fluid permeating the fluid permeable membrane.

34. The method according to claim 33, further comprising:

forming the barrier from a thin film containing at least one of the group consisting of oxides, nitrides, borides, silicides, carbides and aluminides.

35. The method according to claim 34, further comprising:

forming the wire mesh membrane from a stainless steel screen having a mesh count ranging from approximately 19 to 1000 counts per inch.

36. A method of making a fluid separation assembly, comprising:

providing a membrane permeable to a desired fluid and having opposing surfaces;

providing a first retainer adjacent to one of the opposing surfaces of the fluid permeable membrane;

providing a wire mesh membrane having an intermetallic diffusion bonding barrier and adjacent to the other one of the opposing surfaces of the fluid permeable membrane;

providing a permeate member adjacent the wire mesh membrane;

providing a gasket between the fluid permeable membrane and the wire mesh membrane, wherein the periphery of the gasket extends beyond the periphery of the wire mesh membrane;

providing a second retainer between the gasket and the permeate plate; and

hermetically sealing the first retainer, the gasket and the second retainer at their peripheries.

37. The method according to claim 36, further comprising:

forming the barrier from a thin film containing at least one of the group consisting of oxides, borides, silicides, aluminides and nitrides.

38. The method according to claim 36, further comprising:

forming the wire mesh membrane from a stainless steel screen with a mesh count ranging from 19 to 1000 mesh per inch.

39. A method for supporting a fluid permeable membrane, comprising:

providing a membrane that is permeable by a desired fluid and having opposing surfaces; and

providing a wire mesh membrane with an intermetallic diffusion bonding barrier, wherein the wire mesh membrane is adjacent to one of the opposing surfaces of the fluid permeable membrane.

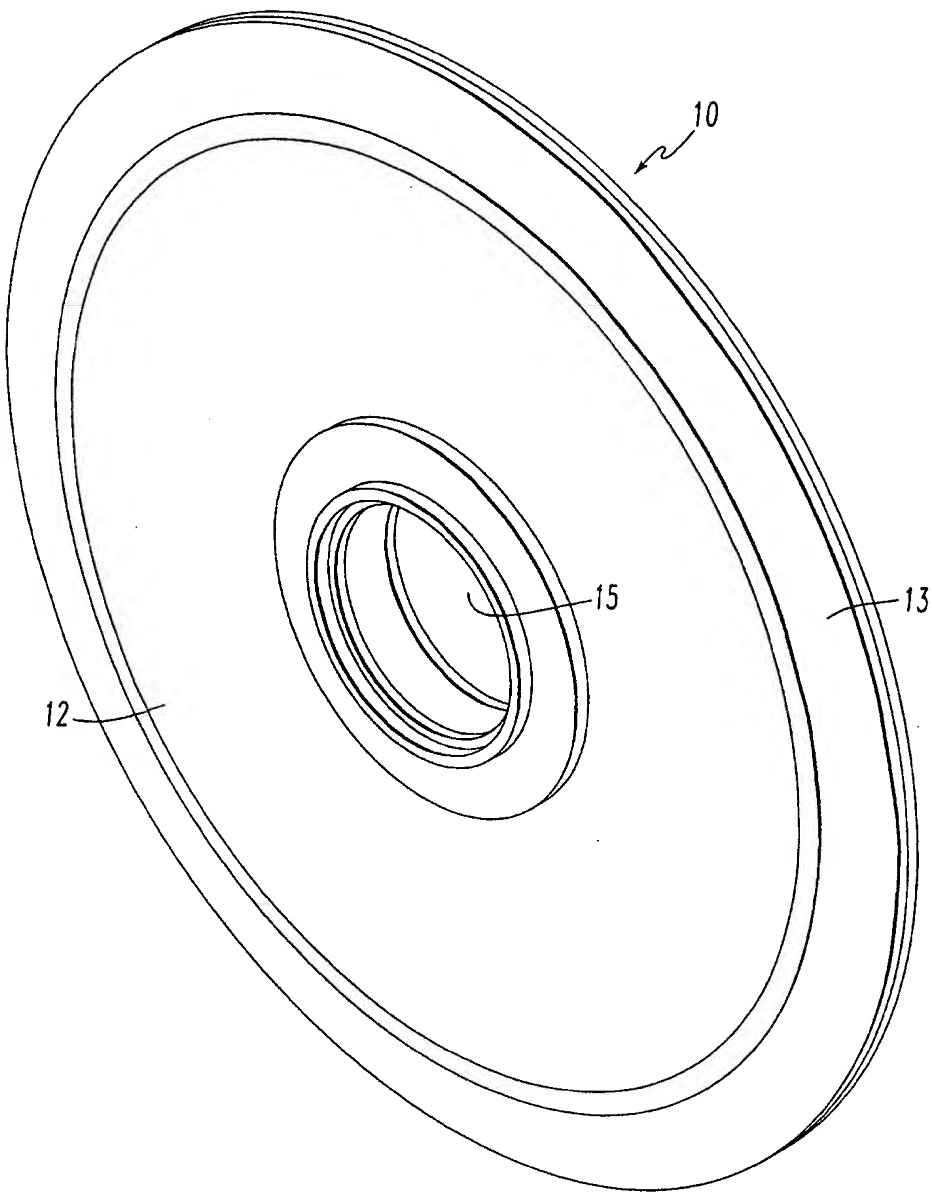



FIG. 1

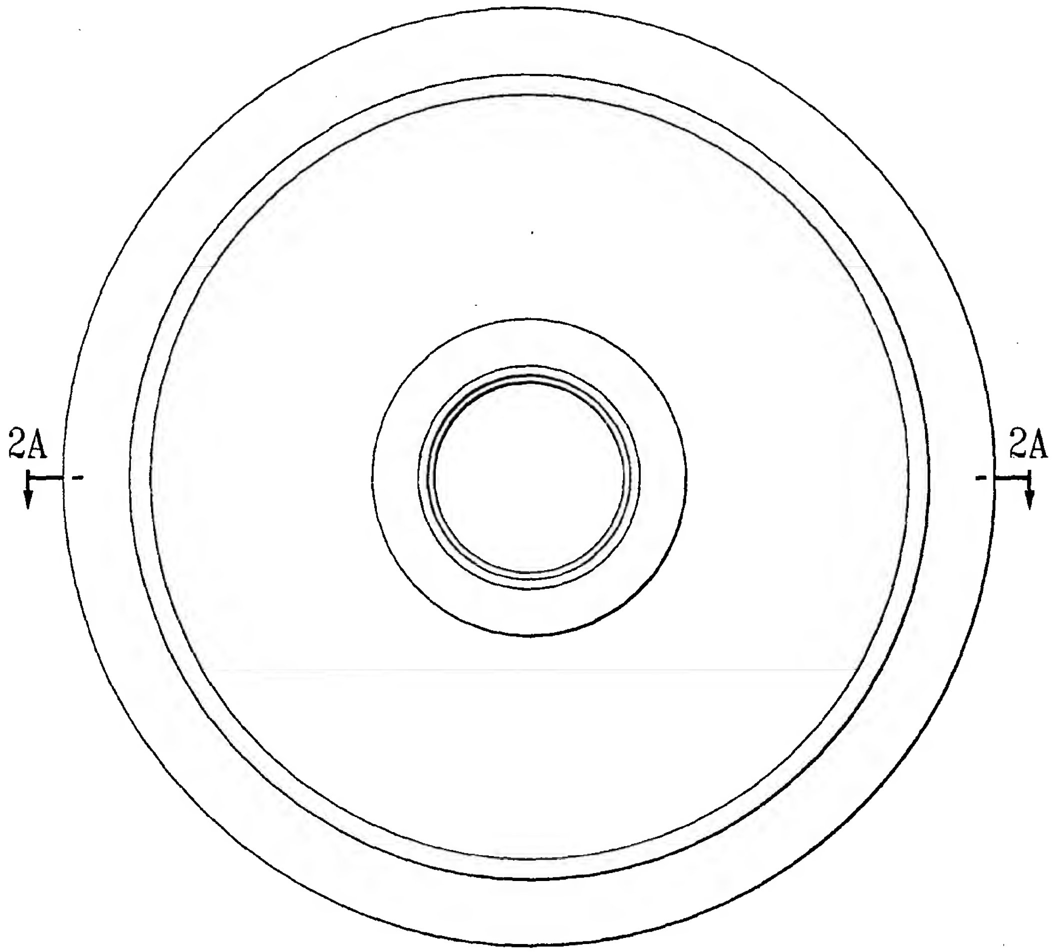



FIG.2

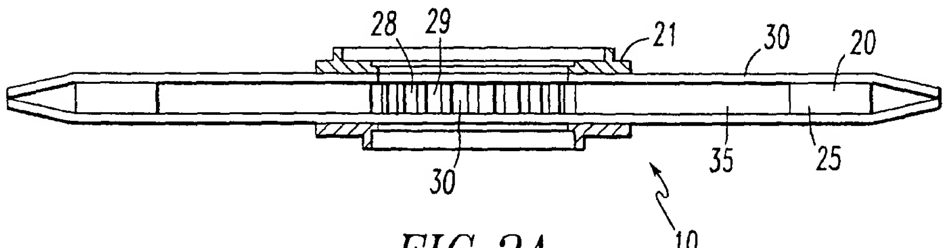



FIG.2A

FIG.3

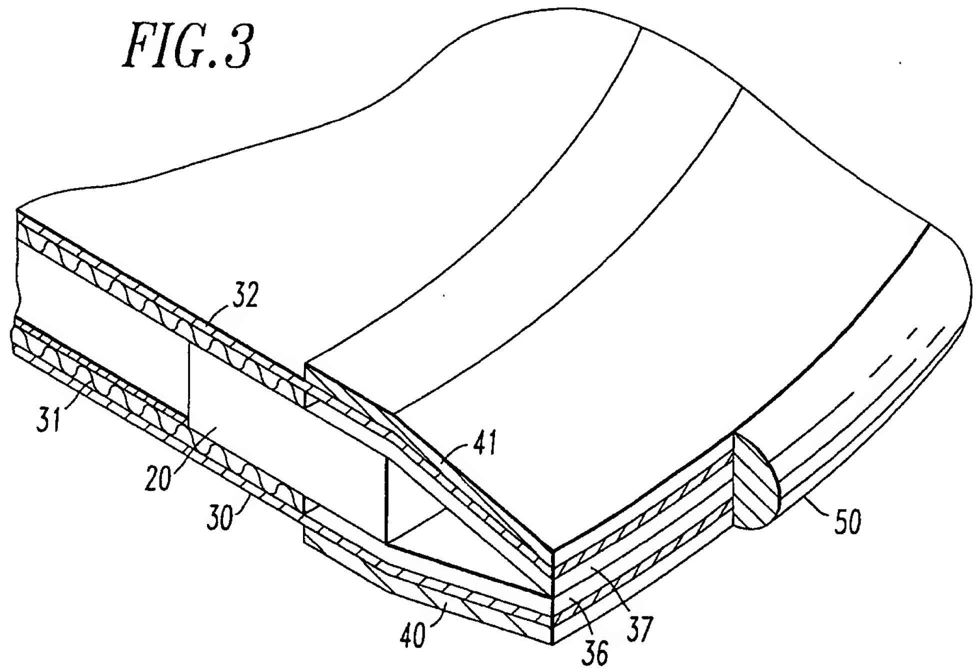



FIG.4

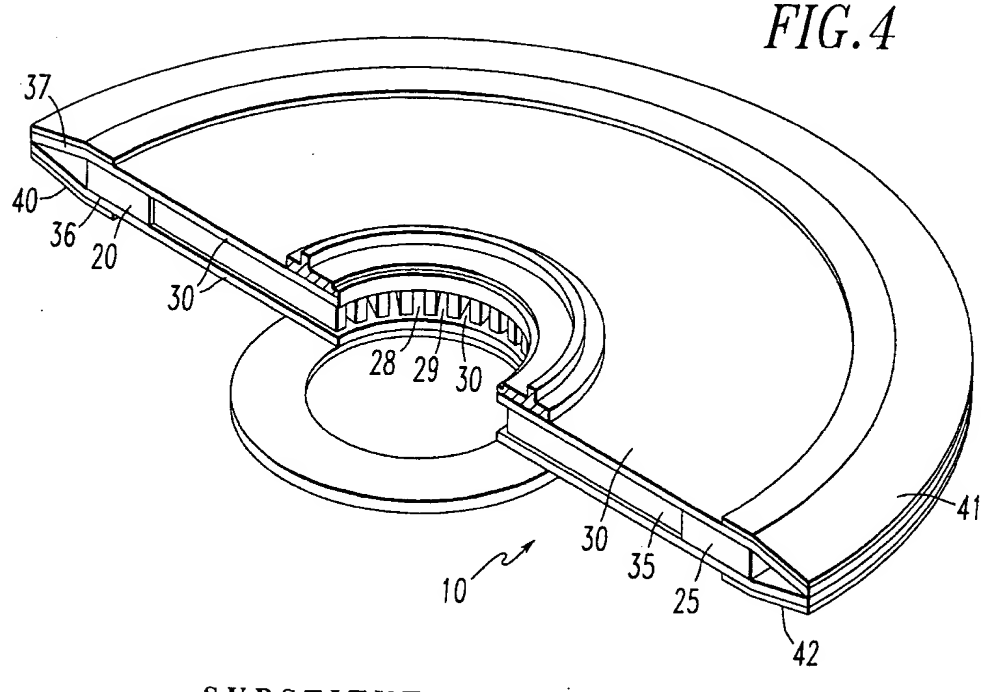
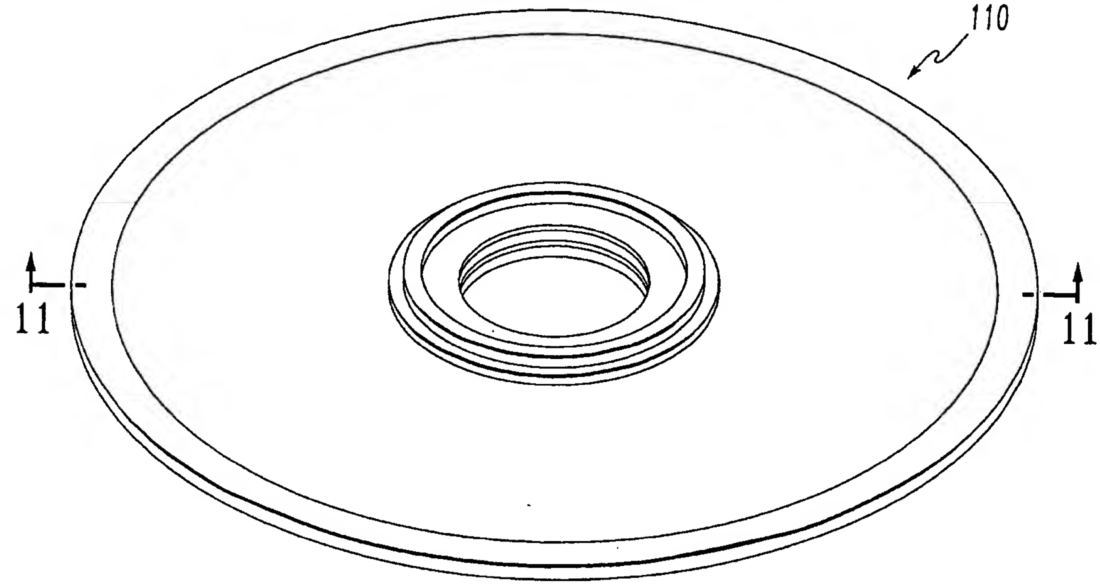


FIG.5



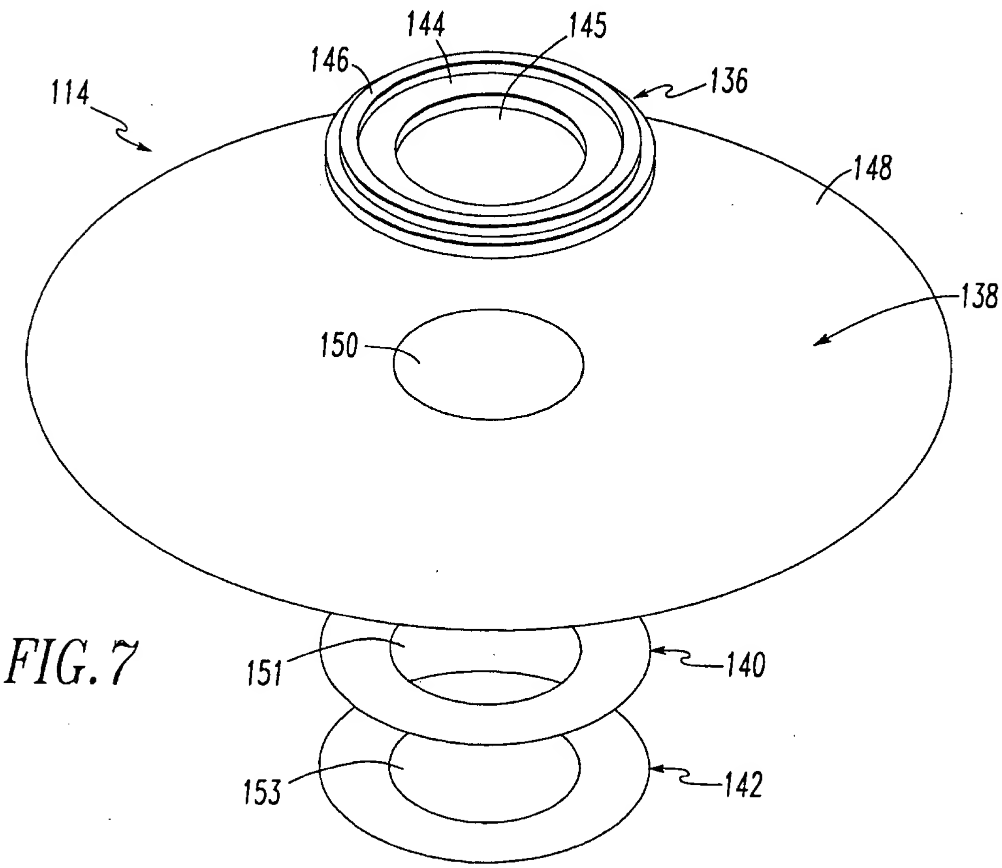




FIG.7

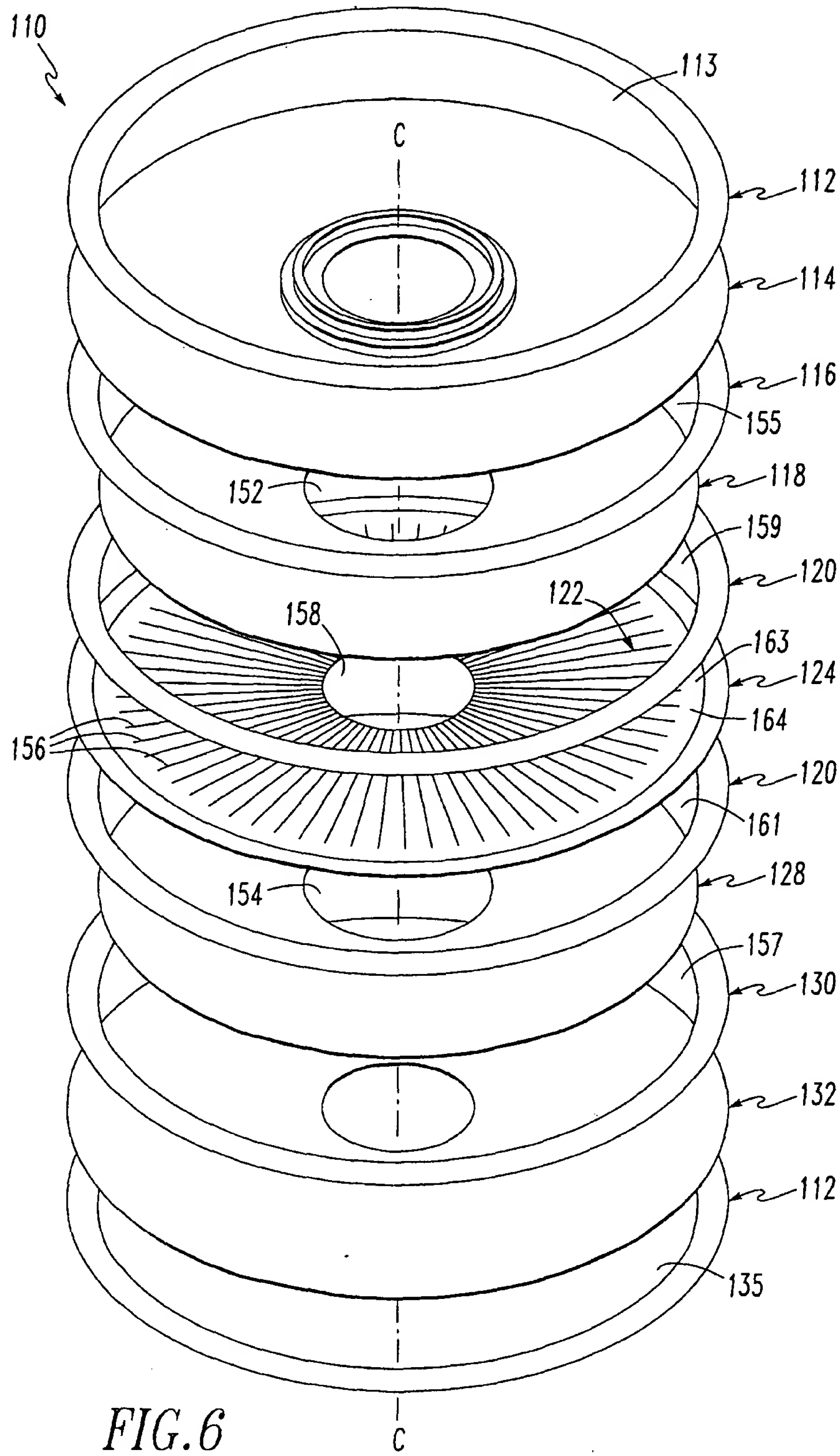

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FIG.6

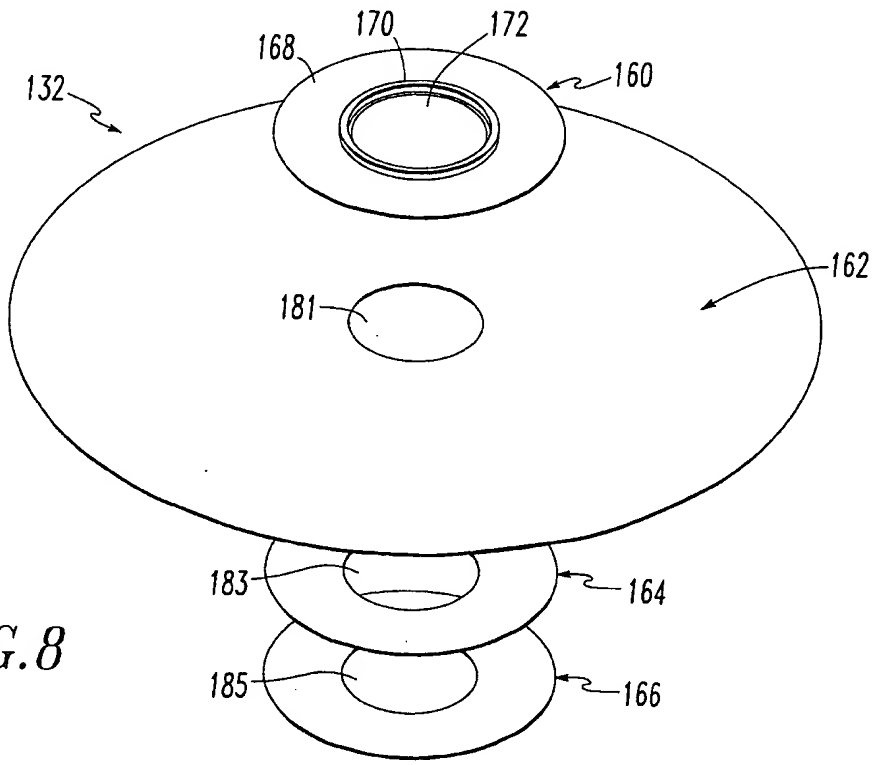


FIG. 8

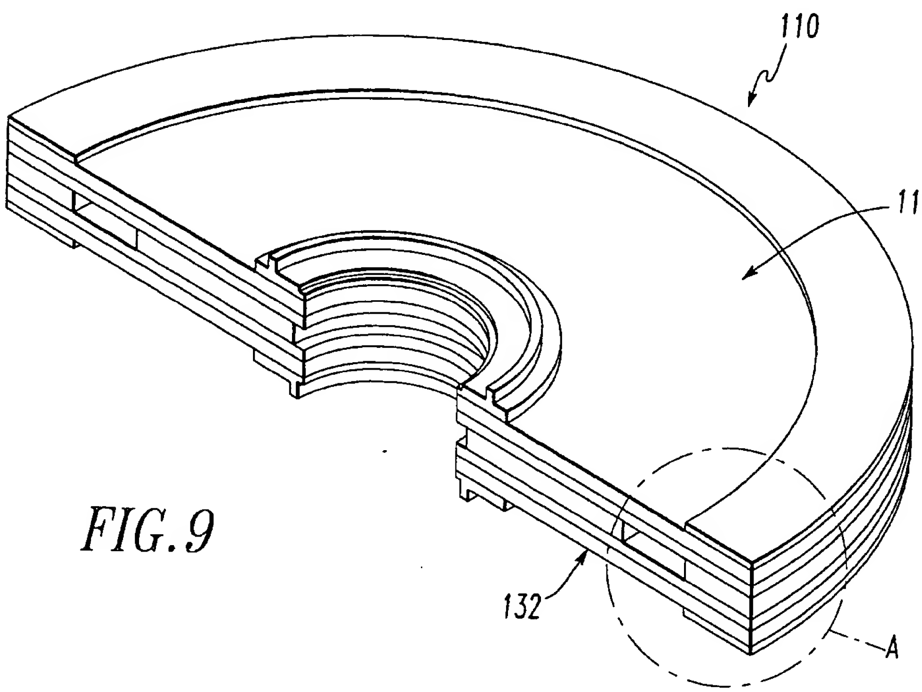


FIG. 9

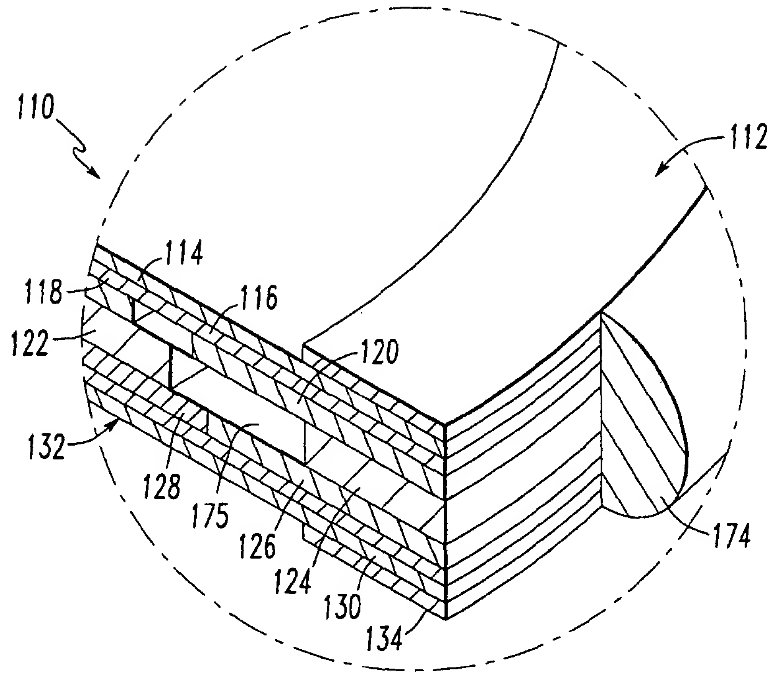
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*FIG.10*

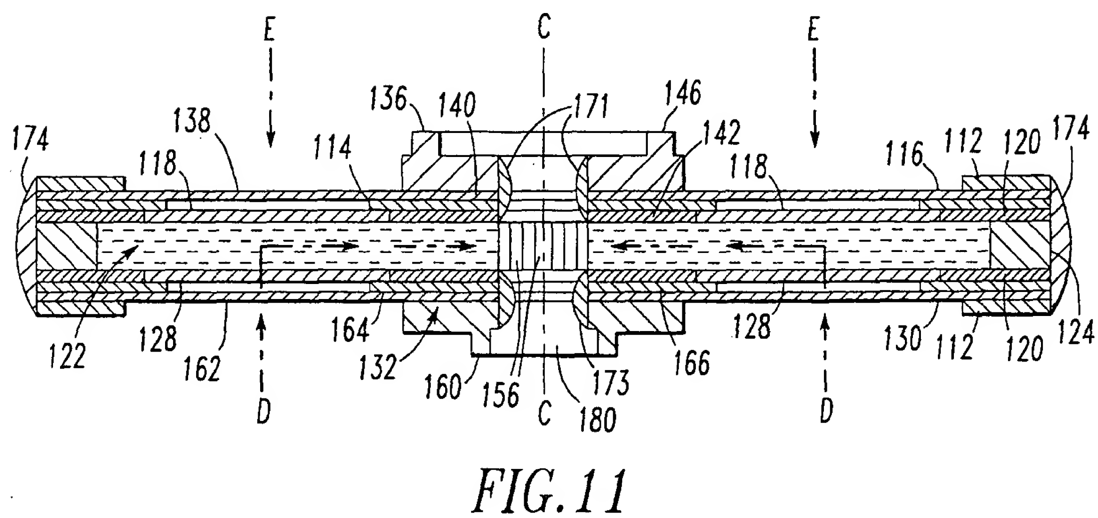
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*FIG.11*

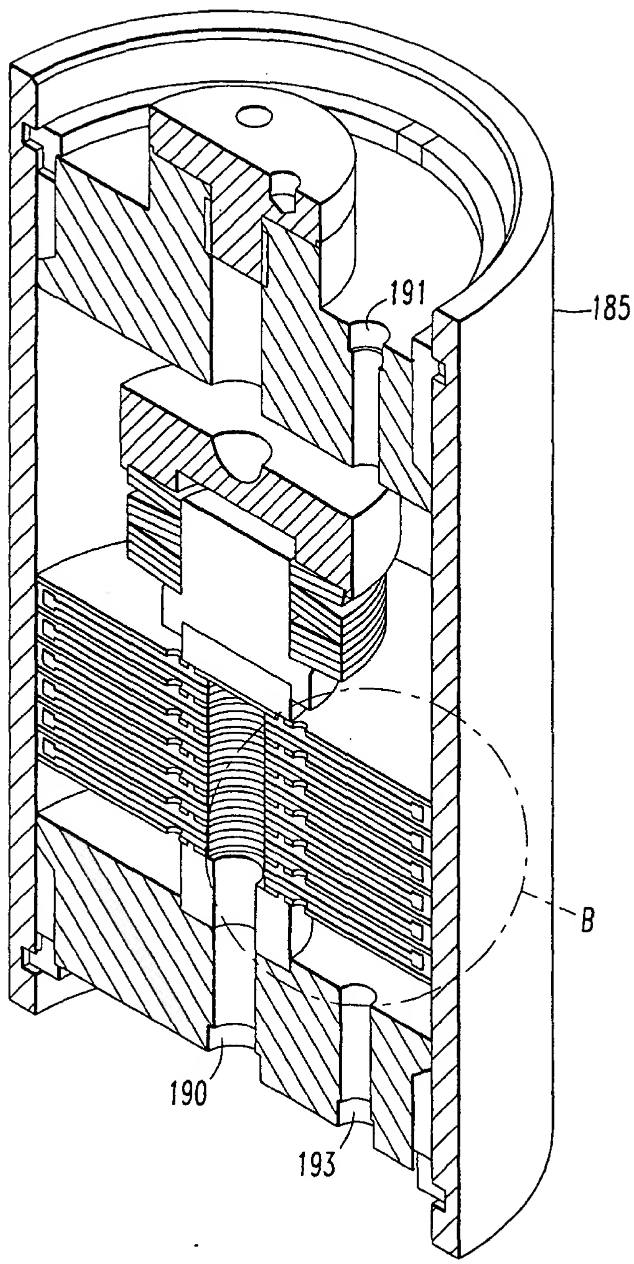


FIG.12

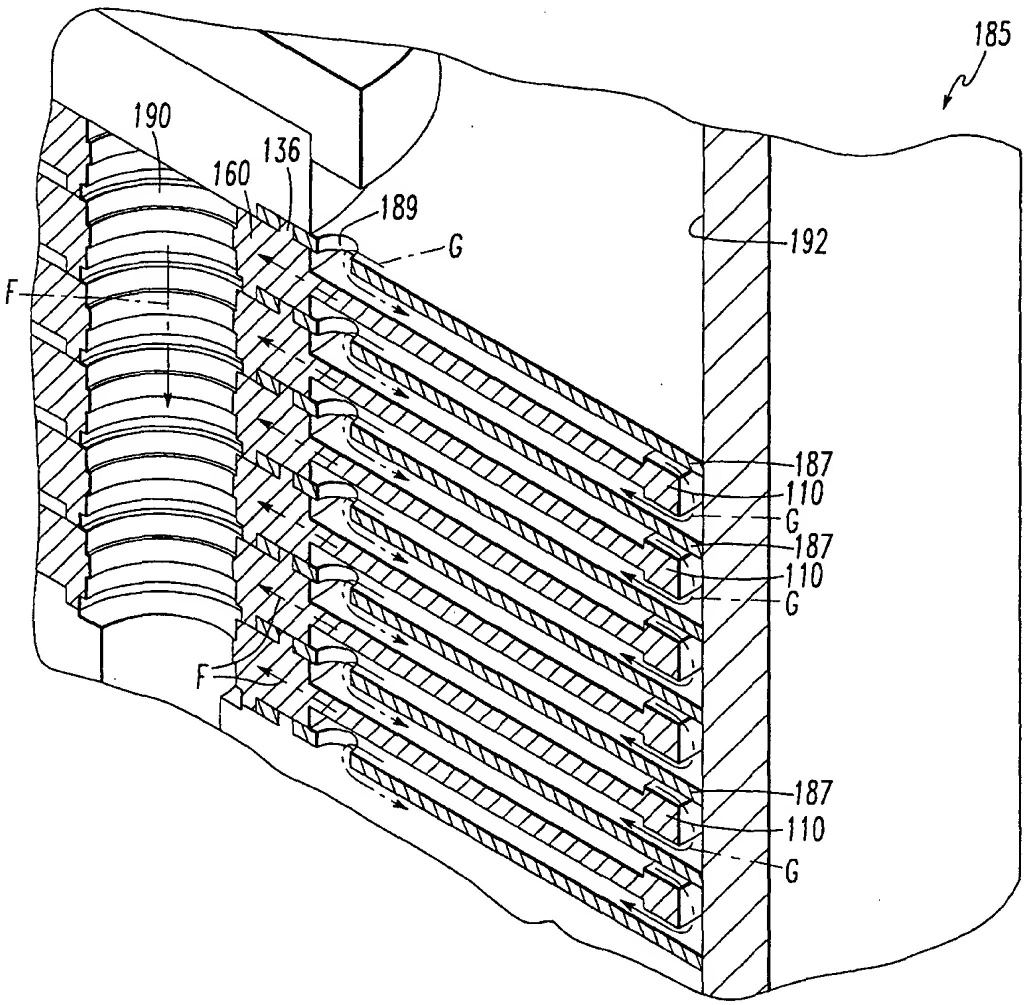


FIG. 13

# INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 99/26527

**A. CLASSIFICATION OF SUBJECT MATTER**
IPC 7 B01D53/22 B01D63/08

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

| Category ° | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
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| A | US 4 699 637 A (INIOTAKIS NICOLAS ET AL)<br>13 October 1987 (1987-10-13)<br>claim 1 | 2,33,39 |

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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"&" document member of the same patent family

| Date of the actual completion of the international search | Date of mailing of the international search report |
|---|---|
| 13 March 2000 | 22/03/2000 |

| Name and mailing address of the ISA | Authorized officer |
|---|---|
| European Patent Office, P.B. 5818 Patentlaan 2<br>NL - 2280 HV Rijswijk<br>Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,<br>Fax: (+31-70) 340-3016 | Faria, C |

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# INTERNATIONAL SEARCH REPORT

Inte ional Application No
PCT/US 99/26527

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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